ISBN: 978-3-95450-208-0 doi:10.18429/JACoW-IPAC2019-WEPRB011

PVD DEPOSTION OF Nb_3Sn THIN FILM ON COPPER SUBSTRATE FROM AN ALLOY Nb_3Sn TARGET

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Abstract

In this study we report on the PVD deposition of Nb_3Sn on Cu substrates with and without a thick Nb interlayer to produce Cu/Nb/Nb_3Sn and Cu/Nb_3Sn multilayer structures. The Nb_3Sn was sputtered directly from an alloy target at room and elevated temperatures. The dependence of the superconducting properties of the total structure on deposition parameters has been determined. The films have been characterized via SEM, XRD, EDX and SQUID magnetometer measurements. Analysis showed that the composition at both room and elevated temperature was within the desired stoichiometry of 24–25 at%. However, superconductivity was only observed for deposition at elevated temperature or post annealing at 650 °C. The critical temperature was determined to be in the range of 16.8 to 17.4 K. In the case of bilayer deposition, copper segregation from the interface all the way to the surface was observed.

INTRODUCTION

Superconducting niobium cavities are used in particle accelerators to provide the gradient required to accelerate the desired particles via RF energy with low losses. Bulk niobium (Nb) has been the material of choice for superconducting RF (SRF) cavities since it has the highest critical temperature (T_c = 9.25 K) and the highest superheating magnetic field H_{sh} of all the pure metals, and also because it can be formed easily into a cavity shape. The RF performance of bulk Nb cavities has continuously improved over the years and is approaching the optimal performance achievable (H_{sh} ~ 210 mT) [1]. Although further improvement has been achieved with N surface doping [2, 3, 4], alternative solutions giving even greater SRF surface efficiency enhancement need to be pursued. The aim is to find an alternative material that can sustain very large surface magnetic fields (i.e. high H_{c2}), as this is what fundamentally limits the accelerated energy E_{acc}, and therefore sets the minimum number of cavities required to reach a given energy.

At ~2 K, cavities made from a material with lower surface resistance R_s than niobium would have smaller losses at a given E_{acc}, so they would allow the cryogenic plant to be smaller and require less grid power, or they could operate at higher gradients, and therefore fewer cavities would be required. Furthermore, the temperature dependent part of R_s scales with $e^{-Tc/T}$, so if the alternative material had a large T_c, low R_s operation could be possible at 4.2 K, greatly simplifying the cryogenic plant by allowing it to operate at atmospheric pressure. Low R_s operation at even higher temperatures would open up the possibility of using helium gas for cooling, giving even greater savings.

The surface resistance R_s is defined as the sum of $Rs = R_{BCS(T)} + R_{res}$ where $R_{BCS} \propto e^{-Tc/T}$ and R_{res} is independent of temperature and is due to parasitic losses. The residual resistance R_{res} is not yet fully understood, but is believed to be significantly due to intrinsic losses caused by imperfect surface quality, metallic inclusions within the penetration depth, the presence of oxides on the surface, and grain boundaries. Other contributors are extrinsic losses due to flux trapped during cooling. Due to the wide variety of phenomena, it is impossible to predict these residual losses with one formula. However, from results reported in literature [5], R_{res} is found empirically to be proportional to $\sqrt{\rho_n}$ where ρ_n is normal state resistivity.

Nb_3Sn is superconductor with T_c = 18 K, ρ_n = 8-20 μΩ·cm and H_{c2} = 28 T. Its critical temperature is higher than that of Nb (9.3 K), and hence, at 4 K, it has an RF resistance an order of magnitude lower than that of Nb and so a superior quality factor. In recent years, there has been an extensive effort, with some degree of success, to convert a Nb cavity into Nb_3Sn by alloying the inner surface of the cavity through Sn diffusion at high temperature. However, the lack of reproducibility remains a major hindering and limiting factor [6].

Hence, we are investigating the properties of deposited Nb_3Sn films. We have characterised the structure, composition and DC superconducting properties of films deposited directly on Cu and on an intermediate Nb layer.

EXPERIMENTAL SETUP

The deposition chamber, described in detail in [7], has ports which allow the installation of up to four magnetrons facing downwards towards the substrate at an angle of 45°. Three 3-inch planar magnetrons were used. One was mounted on an adjustable bellows allowing it to move to between 50 and 250 mm from the substrate surface. The other magnetrons were fixed at 150 mm from the substrate surface. Pressure during deposition was typically 10^{-3} mbar and was set by adjusting the flow rate through the mass flow controller, with a constant pumping speed, until the desired pressure was read from the Baratron.

The sample temperature during deposition was measured using a thermocouple located at the heater filament. Another, retractable, thermocouple was mounted inside the

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deposition chamber and can measure the substrate temperature by direct contact. After each air exposure the chamber was baked to achieve a pressure of $P = 10^{-10}$ mbar.

RESULTS AND DISCUSSION

Nb_3Sn Deposition in Single Layer

Nb_3Sn films were deposited on an oxygen-free, high-conductivity (OFHC) copper substrate without any surface chemical polishing preparation. The deposition was carried out at 650 °C (sample A15-6), 450 °C (A15-7) and at room temperature (A15-9). For the latter, the substrate was heated to 650 °C for 20 hours and then cooled to room temperature prior the deposition and then annealed at 650°C post deposition for 20 hours.

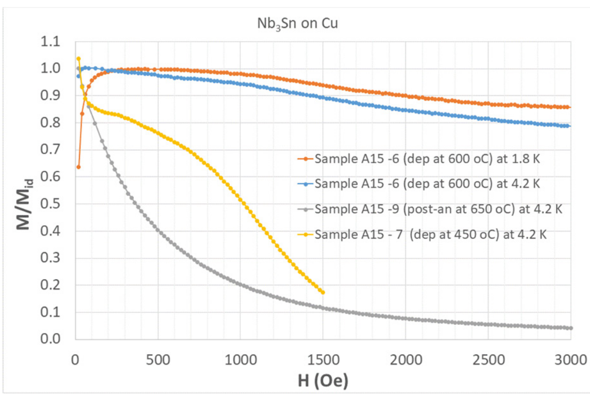

Figure 1: Normalized DC magnetization curves as a function of external applied field for Nb_3Sn film deposited on copper for samples A15-6, A15-7 and A15-9.

Figure 1 shows for normalized magnetic moment (M/M_i) as a function of external field during DC magnetisation measurements. The best performance in terms of superconducting properties was from the film deposited at 650 °C (A15-6) where the film deviated only slightly from the Meissner state even up to a field of 3000 Oe. The film deposited at room temperature (A15-9) showed no superconductivity until it was annealed and then had the worst performance since M/M_i dropped sharply at a very low field of about 100 Oe. The film deposited at moderate temperature of 450 °C (A15-7) performed slightly better, but its performance was still significantly below of that of sample A15-6. This was also evident in the critical temperature, measured as 14.6 K for sample A15-7 and 15.7 K for sample A15-6. Both samples were below the ideal T_c reported for the Nb_3Sn which is 18 K.

The planar and cross-section SEM together with the grazing-incidence X-ray diffraction (GIXRD) (Fig. 2) show that the film of sample A15-6 consists of a compact, dense structure of small grains with a reasonably sharp layer interface.

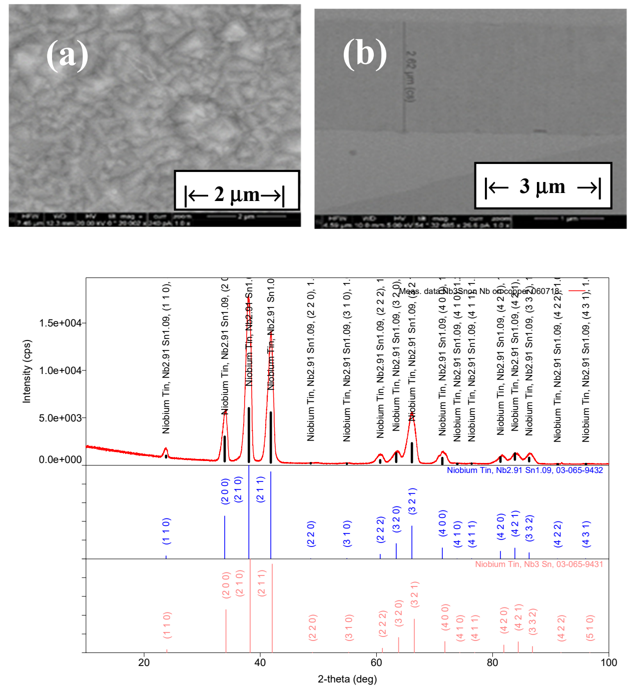

Figure 2: (a) Planar and (b) cross-section SEM micrograph, and c) the GIXRD spectrum of sample A15-6.

The grain size calculated from the X-ray diffraction is in the range of 8 to 10 nm. This is about twice the calculated coherence length of Nb_3Sn, which is reported to be 4.2 nm [7]. The lattice parameter has also been calculated to be 5.292 Å. The Energy Dispersive X-Ray (EDX) analysis showed the film composition was within the tolerance limit of the stoichiometry of the superconducting phase of Nb_3Sn and impurity free within the detection limit. The reduction in T_c can be explained by the lack of long range order as well as a small deviation from the ideal stoichiometry.

Nb_3Sn Deposition in Bilayer

To assess the likely performance of Nb_3Sn deposited on a Nb cavity as an enhancement to a pure Nb cavity, a bilayer of Nb/Nb_3Sn was made. Sample A15-4 was synthesised by depositing Nb_3Sn of top of 2.6 µm of Nb on an untreated OFHC copper substrate at 650 °C. Figure 3 shows that the bilayer deposition resulted in two different and distinct areas. In the homogeneous area as shown in Fig. 3 (a) and (b), the layer has grown in the intended structure. The SEM cross-section shows there are two distinct and well separated layers with sharp interface following contour of the substrate roughness. The planar view also shows a mosaic of a well-defined large grains of Nb_3Sn densely packed at the surface. The scanning EDX line analysis (Fig. 4), shows the purity of the chemical state of the grown bilayer but with some degree of a mixed state at the interface. The latter could be partly due to resolution of the analysis being limited by the sampling volume of the analysing electron beam.

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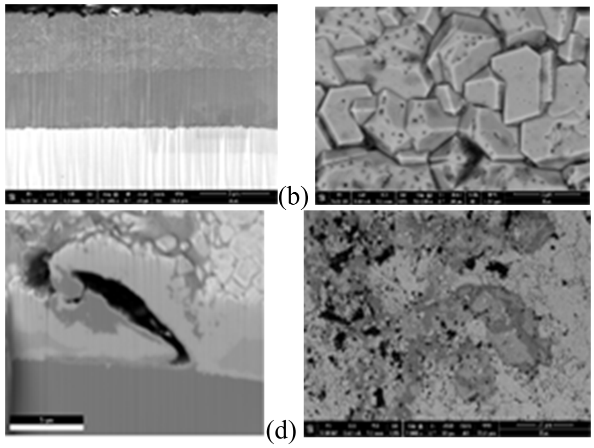

Figure 3: High resolution SEM micrograph of FIB X-section and planar view of sample A15-4 (a)-(b) pure Nb_3Sn area (c)-(d) segregated copper area.

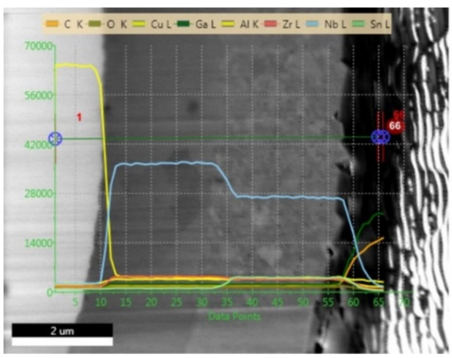

Figure 4: EDX line scan of homogeneous area of sample 15-4.

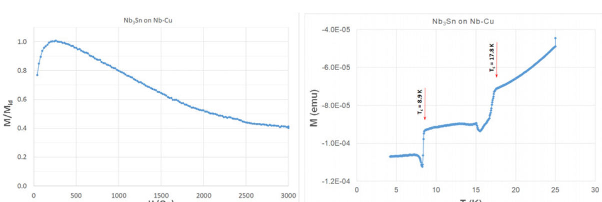

Figure 5: Normalized dc magnetization curves as a function of external applied field (left) and magnetization as a function of temperature (right) for Nb_3Sn/Nb bilayer film deposited on copper for sample A15-4.

One can also observe that level of the Nb counts (Y axis in Fig. 4) drops by ~25% in the Nb3Sn layer. Furthermore the level of other impurities such as oxygen and carbon is in the measurement noise and could be entirely due to surface contamination.

However, as shown in Fig. 3 (c) and (d), in some areas of sample A15-4 the film grew in a very complex structure. From both sets of micrographs (cross section and plan view) one can clearly see the layers of Nb and Nb_3Sn are completely intermixed and there is a substantial volume of copper, both elemental and in the form of copper-tin alloy, present throughout the depth of the layer and at the surface. The elemental mapping determined by EDX analysis of the surface and the cross-section of the area showed that the surface is a mix of copper-tin alloy, Nb_3Sn and copper. A similar mix is also evident starting from the copper-niobium interface and extending all the way up to the surface. Figure 5(a) shows the normalized magnetic moment of the film as function of applied external magnetic field. A drop in M/M_i ratio from unity represents the onset of flux penetration and the deviation from the Meissner state in the superconducting film. In contrast with the single layer of Nb_3Sn deposited in exactly the same conditions, the onset of the drop in magnetisation starts at a much lower field of 500 Oe. The critical temperature of the Nb_3Sn layer is found to be at 17.8 K as shown is in Fig. 5(b) which is much closer to the reported value of 18 K. However, the drop in magnetisation is gradual, which could be due to the presence of mixed phases, an area of Sn deficiency, or some other type of defect such as impurities, dislocations, vacancies or interstitials. The critical temperature of the Nb layer is found to be at 8.9 K which is 0.3 K lower than is usual for thin-film Nb, but the drop in magnetisation is sharp. The GIXRD showed that the diffracted peaks match well with Nb_3Sn stored data. The grain size was calculated to be 120 Å with a lattice parameter of 5.291 Å.

The best superconducting properties among a wider study of deposited Nb_3Sn films were found in the samples deposited on Cu at high temperature (around 550-650 °C). On the other hand, the samples deposited at room temperature show no superconductivity after deposition but showed some superconductivity after being post annealed at 650 °C. In general, Nb_3Sn looks to be a good material for a single-layer coating and the properties should be further tested under RF conditions.

Depositing a bilayer film (first Nb followed by Nb_3Sn) aimed to synthesise a system which is similar to a bulk Nb cavity with a Nb_3Sn layer produced by Sn diffusion process.

However, the EDX and SEM results showed that there were some areas that grew with a non-homogeneous structure. This was caused by diffusion of Cu from the substrate into the bilayer. Such defects will be detrimental to the performance of the RF cavity operating in the RF field. Eliminating this problem needs to be addressed through further research if this technique is to be a viable way of enhancing pure Nb cavities.

CONCLUSION

Nb_3Sn can be successfully deposited from an alloy target and demonstrate good SC properties when it is deposited at high temperature (around 650 °C). Complex defects can be formed in when Nb_3Sn is deposited in a bilayer structure with Nb.

ACKNOWLEDGEMENT

This work was conducted under the aegis of the Science and Technology Facility Council (STFC). The authors would also like to acknowledge the support provided by European Union's H2020 Framework Programmes awarded to the Science and Technology Facilities Council: ARIES collaboration under grant agreement no. 730871.

ISBN: 978-3-95450-208-0 doi:10.18429/JACoW-IPAC2019-WEPRB011

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